Production and Isolation of $^{237}$U and Fission Fragments Resulting from the $^{238}$U($\gamma$, n) and $^{238}$U($\gamma$, f) Reactions

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Objective

Photonuclear Production

- Predictive modeling
- Irradiation techniques
- Identification techniques
- Isotope purification
  - Chemical processes
  - Mass separation

**Demonstrate** $^{237}\text{U}$ and fission fragment production from $^{238}\text{U}(\gamma,n)$ and $^{238}\text{U}(\gamma,f)$ reactions.

Radioisotope Uses

- Tracers
- Nuclear forensics
- Medical isotopes
Photonuclear Production

- Idaho Accelerator Center electron linear accelerator $\rightarrow$ tungsten converter $\rightarrow$ bremsstrahlung

- Bremsstrahlung curve simulated with MCNP6

- Cross sections obtained from the Evaluated Nuclear Data File
  - At the lower end of energies the production of $^{237}$U is more likely

- Accelerator energy increase:
  - greater flux magnitude
  - higher photon endpoint energy
  - photons more forward directed
Predictive Modeling Using MCNP

• Help determine optimal experiment geometries

• Uranium foil fully positioned with in the beam diameter 10 cm from the converter

• Predict sample activity
  – Transport
  – Radiation safety
  – Production Yield
    • (ENDF/B-VII.1 (γ,n) cross section)
    • 1 hr. irradiation
    • 21.5 MeV
    • 0.1 g
    • ~90 μCi · kW⁻¹ · hr⁻¹ · g⁻¹
      • ~50% within (-1,1)
      • ~70% within (-2,2)
      • ~85% within (-3,3)
$^{238}\text{U Sample Irradiation}$

- Optimal accelerator energy
  - Stable 21.5 MeV
  - Endpoint energy beyond peak of cross section.

- Optimal accelerator current
  - Largest achievable

- Experiment parameters
  - 50 $\mu$A
  - 0.1 g $^{nat}$U foil
Identifying $^{237}\text{U}$

**Detection Details**
- 4 hrs. post irradiation
- 15 hr. detection

**Cross Section Details**
- $^{238}\text{U}(\gamma,n)$ Threshold: ~6.2 MeV
- Competes with $^{238}\text{U}(\gamma,f)$

**Identification**
- Observed 4 photo-peaks (FP interferences)
- Data trends to half-life curve

**Production Yield**
- $\sim 98 \pm 0.1 \ \mu\text{Ci} \cdot \text{kW}^{-1} \cdot \text{hr}^{-1} \cdot \text{g}^{-1}$ (9% difference)
Chemical Separation at INL

$^{238}\text{U}, ^{237}\text{U}$ & Fission Products

Step 1: Dissolve Irradiated Foil
- Loading Solution: 10 M HNO$_3$
- Immediate Elution

U & Zr

Step 2: Eichrom® UTEVA Resin
- Loading Solution: 5 M HNO$_3$
- Elution with 0.05 M HNO$_3$

Lanthanides

Step 3: Eichrom® RE Resin
- Elution with 0.05 M HNO$_3$

Non-Lanthanide Fission Products
- $^{99m}\text{Tc}$
- $^{99}\text{Mo}$
- $^{115}\text{Cd}$
- $^{115m}\text{In}$
- $^{133}\text{I}$

Interesting Isotopes
- $^{95}\text{Zr}$ – medical isotope
- $^{99m}\text{Tc}$ – organ imaging
- $^{99}\text{Mo}$ – decays to $^{99m}\text{Tc}$
- $^{115}\text{Cd}$ – trace absorption and excretion of cadmium in tissues
- $^{115m}\text{In}$ – used to evaluate certain diseases
- $^{133}\text{I}$ – used to map brain tumors
- $^{237}\text{U}$ – trace uranium in ground water
Foil Dissolution

Dissolution Details
- ~1 hr.
- 10 M nitric acid
**UTEVA Column**

**Half-lives**
- $^{99}$Mo – 2.8 days
- $^{99m}$Tc – 6.0 hrs
- $^{115}$Cd – 2.2 days

**Detection Details**
- UTEVA rinse
- 27.5 hrs. post irradiation
- 0.7 hr. detection

**Events vs. Energy (keV)**
- $^{153}$Sm 70/103 keV (I=5/29%)
- $^{99m}$Tc 141 keV ($^{99}$Mo $\rightarrow$ $^{99m}$Tc)
- $^{99}$Mo 181 keV (I=6%)
- $^{115m}$In 336 keV ($^{115}$Cd $\rightarrow$ $^{115m}$In)
- $^{115}$Cd 492/528 keV (I=8/27%)
- $^{99}$Mo 366 keV (I=1%)
- (cancer treatment/diagnostics)
Elution from UTEVA Resin

Half-lives

- $^{237}$U – 6.8 days
- $^{97}$Zr – 16.7 hours
- $^{237}$U most abundant radioisotope: ~ 6 days
- 87% of fission products with half-life < 1 day
  - 8% > 6 days

Detection Details

- Elution
- 26.3 hrs. post irradiation
- 30 minute detection
Isotopic Purification

Mass separation

• Isotopes separated with mass separator
• The uranium sample is positively ionized thermally and by electron current
• Ion beam propagates through a bending magnet
• Lighter isotopes are deflected more sharply
Isotopic Purification

Catcher Foils
- Ion beams propagate into catcher foils
- 1% collection efficiency
- $5\times10$ Faraday cup catches charged particles used to determine the number of ions
- Pixel array readout characterizing the beam profile
Conclusions & Future Agenda

• Demonstrated photonuclear production of:
  – $^{237}\text{U}$, $^{99m}\text{Tc}$, $^{99}\text{Mo}$, $^{115}\text{Cd}$, $^{115m}\text{In}$, $^{133}\text{I}$, $^{153}\text{Sm}$

• Optimize production and increase activities
  – Modify accelerator parameters

• Optimize sample transport
  – Goal under 3.5 hrs. for packaging and transport

• Optimize separation for isotopes of interest
  – Separation goals:
    • Uranium $\rightarrow$ 2.5 hrs.
    • Non-lanthanides in 3 hrs.
      – Strategy to optimize for each isotope of interest
    • Lanthanides $<$ 7.5 hrs.

• Continue mass separator development
  – Optimize uranium separation
  – Introduce new radioisotopes
Thank you.